

# Best Available Copy

Europäisches Patentamt



(19) European Patent Office

Office européen des brevets



(11) Publication number: 0 555 186 A1

(12)

## EUROPEAN PATENT APPLICATION

(21) Application number: 93810063.3

(51) Int. Cl.<sup>5</sup>: B27K 3/34

(22) Date of filing: 02.02.93

(30) Priority: 05.02.92 GB 9202378

(72) Inventor: McDade, Mark Daniel  
428 Utica Avenue  
Boulder, Colorado 80304 (US)

(43) Date of publication of application:  
11.08.93 Bulletin 93/32

(84) Designated Contracting States:  
AT BE DK ES GR IE LU NL PT SE

(71) Applicant: SANDOZ LTD.  
Lichtstrasse 35  
CH-4002 Basel (CH)

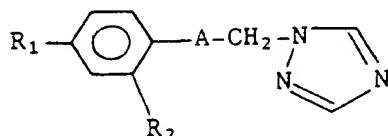
(84) BE DK ES GR IE LU NL PT SE

(71) Applicant: SANDOZ ERFINDUNGEN  
VERWALTUNGSGESELLSCHAFT M.B.H.  
Brunner Strasse 59  
A-1235 Vienna (AT)

(84) AT

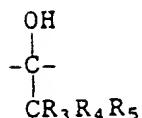
(54) Fungicidal compositions.

(57) Wood preserving composition comprising a compound of formula I

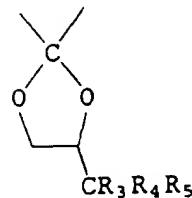


wherein  
A is selected from (i)

XAM

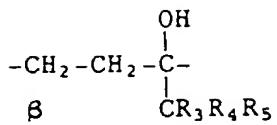


(ii)



and (iii)

EP 0 555 186 A1



whereby the  $\beta$ -carbon attaches to benzene ring of formula (I) ;

$\text{R}_1$  and  $\text{R}_2$  are independently H or Cl ;

$\text{R}_3$  and  $\text{R}_4$  are independently H or  $\text{CH}_3$  ; and

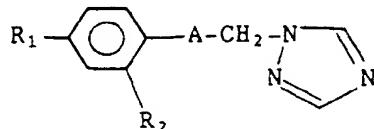
$\text{R}_5$  is methyl, ethyl or cyclopropyl

and method for preserving wood with the aid of a compound of formula (I).

This invention relates to a wood preservative composition and, more specifically, to a wood preservative composition containing a triazole fungicide as active ingredient.

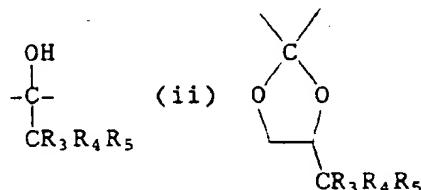
Wood is an important resource material in the construction and industries. Wood can, however, be susceptible to mold, decay and discoloring due to fungal attack. Various compositions are known for combatting such fungal attacks, including certain triazole compounds such as those disclosed in European Patent Application 0 131 684.

It has now been found that certain triazole compounds of the formula (I)

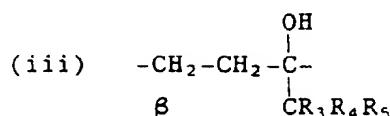


15 wherein

A is selected from (i)



and



35 whereby the  $\beta$ -carbon attaches to benzene ring of formula (I);

$\text{R}_1$  and  $\text{R}_2$  are independently H or Cl;

$\text{R}_3$  and  $\text{R}_4$  are independently H or  $\text{CH}_3$ ; and

$\text{R}_5$  is methyl, ethyl or cyclopropyl.

40 are particularly effective at combatting various fungi which are known to cause mold, decay and discoloration of wood.

Wood, as used herein, refers to any type of wood material or wood product such as plywood, pressed wood, particle-board, wood chip, pulp or intermediates obtained in papermaking.

Particularly preferred compounds of the formula (I) are those in which  $\text{R}_1$  is Cl,  $\text{R}_2$  and  $\text{R}_3$  are H,  $\text{R}_4$  is  $\text{CH}_3$  and  $\text{R}_5$  is cyclopropyl and A is the moiety (i) (commonly known as cyproconazole); those in which  $\text{R}_1$  is Cl,  $\text{R}_2$  is H,  $\text{R}_3$ ,  $\text{R}_4$  and  $\text{R}_5$  are  $\text{CH}_3$  and A is the moiety (iii) (commonly known as tebuconazole); and those in which  $\text{R}_1$  and  $\text{R}_2$  are Cl,  $\text{R}_3$  and  $\text{R}_4$  are H,  $\text{R}_5$  is ethyl and A is the moiety (ii) (commonly known as propiconazole).

The specific compounds mentioned in the preceding paragraph are commercially available. Other compounds falling under the scope of formula (I) are obtainable according to procedures analogous to those known for preparing the commercially available compounds.

50 The compounds of formula (I) for use as wood preservatives are conveniently formulated into compositions comprising a wood preserving or fungicidally effective amount of the compound of formula (I) and an environmentally acceptable carrier for such usage.

The term carrier as used herein means any environmentally acceptable liquid or solid material which may be added to the active constituent to bring it in an easier or improved applicable form, respectively to a usable or desirable strength of activity. It can for example be calcium, magnesium carbonate, xylene or water.

55 The compositions may also be in the form of dispersible powders or granules and will conveniently comprise a surfactant, e.g. a wetting or dispersing agent to facilitate dispersion in liquids of the powder or granules which may contain also fillers and suspending agents.

The aqueous dispersions or emulsions may be prepared by dissolving the active ingredient in an organic solvent optionally containing wetting, dispersing or emulsifying agents and then adding the mixture to water which may also contain one or more surfactants, such as wetting, dispersing or emulsifying agents. Suitable organic solvents are ethylene dichloride, isopropyl alcohol, propylene glycol, diacetone alcohol, toluene, kerosene, methylnaphthalene, polyethyleneglycol, N-methyl-2-pyrrolidone, mixtures of C9 to C11 fatty alcohols, the xylenes, trichloroethylene, furfuryl alcohol, tetrahydrofurfuryl alcohol and glycol ethers.

Typically, the compositions will be in the form of liquid preparations for use as dips or sprays which are generally aqueous dispersions or emulsions containing the active ingredient in the presence of one or more surfactants e.g. wetting agents, dispersing agents or emulsifying agents. The surfactants may be cationic, anionic or non-anionic, all of which are known in the art.

Suitable anionic agents are soaps, salts of aliphatic monoesters of sulphuric acid and salts of sulphonated aromatic compounds.

Suitable non-ionic agents are the condensation products of ethylene oxide with fatty alcohols or with alkyl phenols. Other non-ionic agents are the partial esters derived from long chain fatty acids and hexitol anhydrides, the condensation products of partial esters with ethylene oxide and the lecithins.

The compositions of the invention may contain further adjuvants including thickening agents, antifoam agents, antifreeze agents and suspending agents.

Suitable suspending agents are hydrophilic colloids and vegetable gums.

The compositions for use as aqueous dispersions or emulsions are generally supplied in the form of a concentrate containing a high proportion of the active ingredient, the concentrate to be diluted with water before use. The concentrates may conveniently contain up to 95%, suitably 10-85%, for example 25-60% by weight of the active ingredient. After dilution to form aqueous preparations, such preparations may contain varying amounts of the active ingredient depending upon the type of wood to be treated and the type of fungus, but typically the aqueous preparation will contain from 0.0001% to 10% by weight active ingredient, more typically from 0.001% to 1%.

Methods of applying the compounds to the wood to be treated, such as spraying, dipping, by paint brush, etc., are known to those skilled in the art. Application can be repeated, as necessary.

The formulations listed below are representative of suitable formulations for use in the invention, and are admixed and agitated in accordance with conventional methods to obtain a wood preservative composition.

<u>Formulation 1</u>	
400 g/l	cyproconazole
55 g/l	nonionic polymeric emulsifier blend (e.g. polyalkylene glycol ether/polyoxyethylene alkylaryl ether blend)
66 g/l	antifreeze (e.g. 1,2 propanediol)
3 g/l	thickening agent (e.g. xanthane gum)
1 g/l	bactericide
4 g/l	antifoam agent (e.g. silicon)
balance	water

<u>Formulation 2</u>	
100 g/l	cyproconazole
57 g/l	emulsifier (e.g. a nonylphenolethoxyphosphate)
96 g/l	solvent (e.g. N-methyl-2-pyrrolidone)
balance	solvent (e.g. polyethyleneglycol)

<u>Formulation 3 (emulsifiable concentrate)</u>	
5	100 g/l cyproconazole
	74 g/l emulsifier (e.g. nonylphenyl-hydroxypoly(oxyethylene)phosphate)
	92 g/l emulsifier (e.g. alkyl hydroxypoly(oxyethylene)phosphate)
10	46 g/l solvent (e.g. hexanol)
	101 g/l solvent (e.g. N-methyl-2-pyrrolidone)
	balance solvent (e.g. mixture of C9 to C11 fatty alcohols)

15 **Formulation 4 (wettable granule)**

10 % cyproconazole  
 15 % dispersing agent (e.g. sodium lignin sulfonate)  
 75 % carrier (e.g. calcium magnesium carbonate)

20 **Test of activity against wood destroying fungi in vitro**

25 Suspensions containing a test compound of formula I are incorporated into potato dextrose agar (PDA) to produce a series of five concentrations containing 100 ppm, 10 ppm, 1 ppm, 0.1 ppm, 0.01 ppm resp. of active ingredients. The thus obtained agar test compositions are poured into 9-cm petri dishes. After solidification of the medium, each dish is inoculated with a mycelial disc (5 mm diameter) taken from the periphery of actively growing colonies on PDA (three replicate dishes per isolate per concentration). After incubation (24°C in darkness, 5-14 days depending on the growth rate of the fungi), colony radii are measured. Percentage growth inhibition is calculated on the basis of treated control plates. The EC90 (effective concentration causing 90 % growth inhibition) is determined on the basis of dose-response curves.

30 The compounds of formula (I) are effective in combatting various type of fungi including the following fungi and the symptoms to which they lead.

	<u>Fungus class</u>	<u>Species</u>	<u>Sympton</u>
35	ascomycetes	Sydowia polyspora	dieback/pine
		ceratocysti fagacearum	wilt/oak
40		ceratocysti pilifera	blue stain
		Cephaloascus fragrans	mold
		Physalospora rhodina	discoloration
45	basidiomycetes	Coriolus versicolor	decay
		Poria placenta	decay
		Lentinus lepideus	decay
50		Trametes versicolor	decay

	Serpula lacrymans	mold
	Coniophora putanea	decay
	Gloeophyllum trabeum	decay
5	deuteromycetes	
	Aspergillus niger	discoloration
	Phialophora fastigiata	discoloration
	Alternaria alternata	discoloration
10	Rhinocladiella atrovirens	discoloration
	Gliocladium roseum	mold
	Aureobasidium pullulans	discoloration
15	Trichoderma viride	decay
	Sphaeropsis sapinea	dieback/conifers
	Pencillium expansum	mold

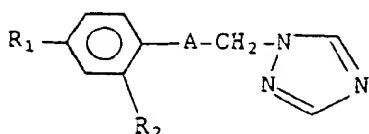
20 **Fungicidal activity**

The compounds cyproconazole, propiconazole and tebuconazole when tested against a variety of fungal diseases demonstrate particularly good activity against basidiomycetes including the fungi Coriolus versicolor, Poria placenta, Serpula lacrymans, Coniophora puteana, Gloeophyllum trabeum, Lentinus lepideus and Trametes versicolor.

Cyproconazole is particularly effective against Poria placenta, Lentinus lepideus and Trametes versicolor.

30 **Claims**

1. A wood preserving composition comprising a wood preserving effective amount of a compound of the formula (I)



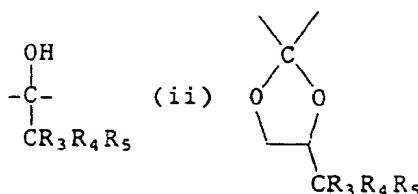
40

wherein

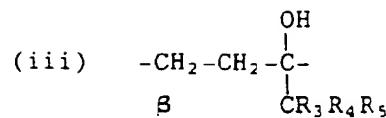
A is selected from (i)

45

50



55 and



whereby the  $\beta$ -carbon attaches to benzene ring of formula (I); the  $\text{R}_1$  and  $\text{R}_2$  are independently H or Cl;  $\text{R}_3$  and  $\text{R}_4$  are independently H or  $\text{CH}_3$ ; and  $\text{R}_5$  is methyl, ethyl or cyclopropyl; and an environmentally acceptable carrier.

- 10
2. The composition of Claim 1 wherein compound of formula I is selected from cyproconazole, propiconazole and tebuconazole.
  3. The composition of Claim 2, wherein the compound of formula I is cyproconazole.

15

  4. The composition of Claims 1 to 3 comprising additionally a surfactant.
  5. A method for preserving wood comprising applying to the surface of said wood a wood preserving effective amount of the compound of Claims 1-3.

20

25

30

35

40

45

50

55

THIS PAGE BLANK (USPTO)



European Patent  
Office

## EUROPEAN SEARCH REPORT

Application Number

EP 93 81 0063

DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int. Cl.5)		
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)		
X	EP-A-0 050 738 (BASF) * claims * ---	1,2,4,5	B27K3/34		
X	US-A-4 542 146 (J. VAN GESTEL TE AL.) * claims *	1,2,4,5			
X	EP-A-0 148 526 (JANSSEN PHARMACEUTICA) * page 10; example 16 *	1,2,4,5			
X	EP-A-0 287 346 (E.I. DU PONT DE NEMOURS) * claims 1,23,28,29 *	1-4			
X	GB-A-2 136 423 (SANDOZ) * claims *	1-4			
A	EP-A-0 458 061 (DESOWAG) ---				
A	EP-A-0 458 060 (DESOWAG) -----				
			TECHNICAL FIELDS SEARCHED (Int. Cl.5)		
			B27K		
The present search report has been drawn up for all claims					
Place of search	Date of completion of the search	Examiner			
THE HAGUE	08 APRIL 1993	DALKAFOUKI A.			
CATEGORY OF CITED DOCUMENTS					
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document					
T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document					

**THIS PAGE BLANK (USPTO)**

**This Page is Inserted by IFW Indexing and Scanning  
Operations and is not part of the Official Record**

**BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images include but are not limited to the items checked:

- BLACK BORDERS**
- IMAGE CUT OFF AT TOP, BOTTOM OR SIDES**
- FADED TEXT OR DRAWING**
- BLURRED OR ILLEGIBLE TEXT OR DRAWING**
- SKEWED/SLANTED IMAGES**
- COLOR OR BLACK AND WHITE PHOTOGRAPHS**
- GRAY SCALE DOCUMENTS**
- LINES OR MARKS ON ORIGINAL DOCUMENT**
- REFERENCE(S) OR EXHIBIT(S) SUBMITTED ARE POOR QUALITY**
- OTHER:** \_\_\_\_\_

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning these documents will not correct the image problems checked, please do not report these problems to the IFW Image Problem Mailbox.**

**THIS PAGE BLANK (USPTO)**